Electronic Supplementary Information

Selective Two-Photon Absorption in Carbon Dots: an extra piece of the puzzle of photoluminescence emission

Carla, I. M. Santos,[a,b*] *Inês F. A. Mariz*,[a] , *Sandra N. Pinto*,[a,c] *Gil Gonçalves*,[d] *Igor Bdikin*, [d] *Paula A.A.P. Marques*,[d] *Maria Graça P.M.S. Neves*,[b] *José M. G. Martinho*, [a] *and Ermelinda M. S. Maçôas*.[a*]

[a] CQFM, Centro de Química-Física Molecular, IN—Institute of Nanosciences and Nanotechnology and CQE, Centro de Química Estrutural, Instituto Superior Técnico,1049-001 Lisboa,

[b] QOPNA, Chemistry Department, University of Aveiro, Campus Universitario de Santiago, 3810-193 Aveiro, Portugal

[c] iBB-Institute for Bioengineering and Biosciences, Department of Bioengennereing, Instituto Superior Técnico, Universidade de Lisboa, Av. Rovisco Pais 1049-001 Lisboa, Portugal.

[d] TEMA-Nanotechnology Research Group, Mechanical Engineering Department, University of Aveiro, Campus Universitario de Santiago, 3810-193 Aveiro, Portugal

Email: cims@ua.pt (Carla Santos); ermelinda.macoas@tecnico.ulisboa.pt (Ermelinda Maçôas)

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Table S1. Survey of the literature reporting two-photon absorption cross-section values. A description of the starting material and the absorption spectrum is given together with the maximum emission wavelength (λ_{em}), emission quantum yield (ϕ_F), width (W) and height (H) of the dots and their reported two-photon absorption cross-section (TPA).

Type of Cdots	Starting material and procedure in brief	ref	Abs (nm)	λ_{em} (nm)	ϕ_F (%)	Width and Height (nm)	TPA (10^3 GM)
N-doped crystalline	GO and DMF (200ºC, 4.5h)	[1]	unstructured (PLEx 370 nm)	520	31	2-3 (W) 1.5 (H)	48
N-doped crystalline	GO and DMF (250ºC, 6h)	[2]	Not reported	550	32	2-3(W)	40
N-doped crystalline	laser ablation of a carbon target in water vapor and post-functionalization with PEG	[3]	unstructured	480	4-10	9 (W) <5 (H)	39
N-doped amorphous	Citric acid and hyperbranched poly-(amino amine) (HPAA) (200ºC, 3h)	[4]	unstructured with peaks at 285 nm and 370 nm	450	17	10 (W)	16
N-doped	Melamine and glycerol in acidic solution (270ºC, 15 min).	[5]	unstructured	400-500	5-22	0.8-1.5 (W)	1.3-2.0
g-C_3N_4	Acid treatment of bulk g-C_3N_4	[6]	unstructured	406	-	4 (W) 0.35 (H)	30
Undoped	Graphite, Hummers method	[7]	unstructured	550	5.8×10^{-4} [a)]	40 (W)	31 [a)]
Undoped crystalline	Oxidation of graphite nanoparticles by Hummers method	[8]	unstructured with shoulder at 260 nm (PLex 300 nm)	520	1.2	3 (W) 0.8 (H)	2.5 [b)]
Undoped	Citric acid (200ºC for 30 min)	[9]	Band centered at 360 nm	465	8.2	10 (W)	0.022
Undoped	Carrot juice (180ºC for 5 h)	[10]	Band at 286 nm	410	11.5	2-7(W)	0.019
S, Se co-doped crystalline	Hydrothermal treatment of polythiophene and diphenyl diselenide in NaOH	[11]	Band centered at 526 nm	730	0.2	20 (W) 4(H)	30
Undoped GO sheets	Exfoliation of graphite by hummers method	[12]	Not reported	550	34	large sheet	47 [c)]

[a)] The quantum yield value is the two-photon emission quantum yield, the absorption cross-section was estimated based on the values of the $\beta(\lambda)$=0.025cm/GW at 800 nm and concentration 0.2 mg/mL [b)] $\beta(\lambda)$= 0.01cm/GW at 800 nm, concentration 1mg/mL [a)] and [c)] large dots and GO sheets can have a high TPA due to a high number of embedded sp^2 domains.

Table S2 - Atomic percentage from the high resolution XPS.

	%C	%O	%N	C/O	N/C	N_{amine}/N_{amide}
GO	0.61	0.39	-	1.55	-	-
b1	0.63	0.33	0.04	1.90	0.07	1.2
b2	0.60	0.31	0.10	1.93	0.16	1.1
b3	0.64	0.28	0.08	2.31	0.13	1.2

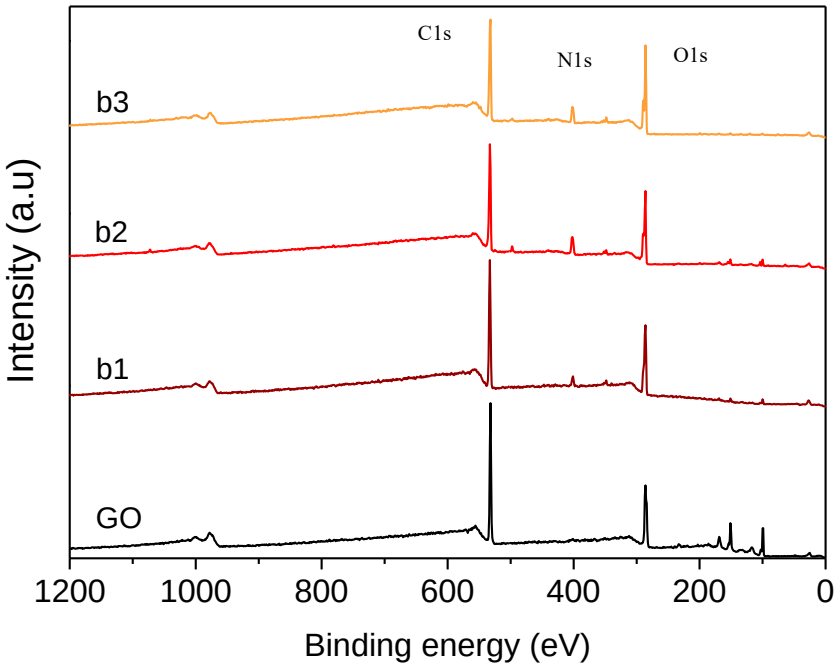

Figure S1. XPS survey spectra of the different batches of nitrogen doped Cdots shown in comparison with the GO spectrum. Note that the N1s peak observed on the N-functionalized dots is absent from the XPS spectrum of GO

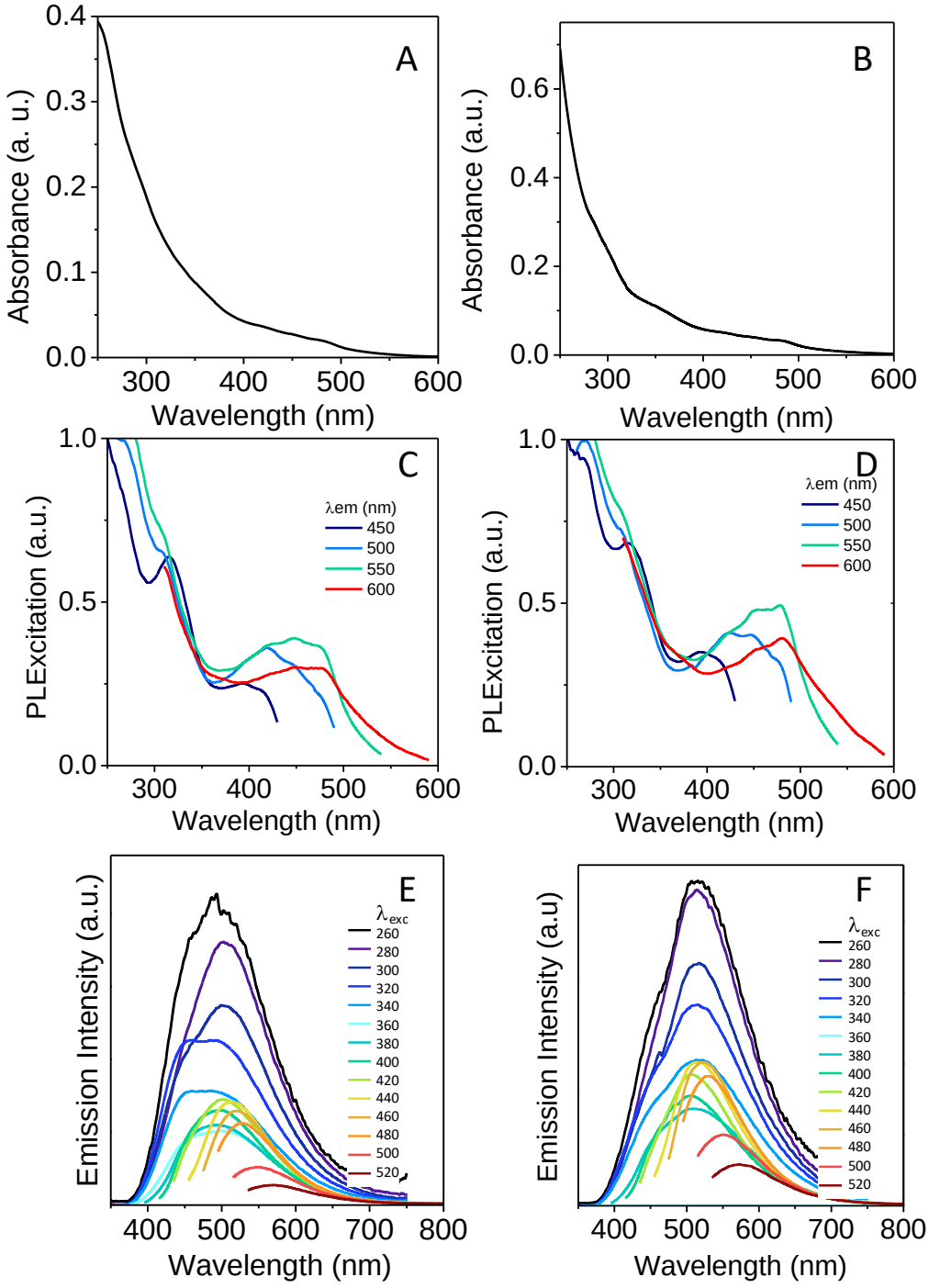

Figure S2. Optical properties of the Cdots in b1 (left) and b2 (right). (A and B) absorption spectra of b1 and b2, respectively. (C and D) photoexcitation spectra collected at different emission wavelength (λ_{em}). (E and F) Emission spectra excited at different wavelengths (λ_{exc}).

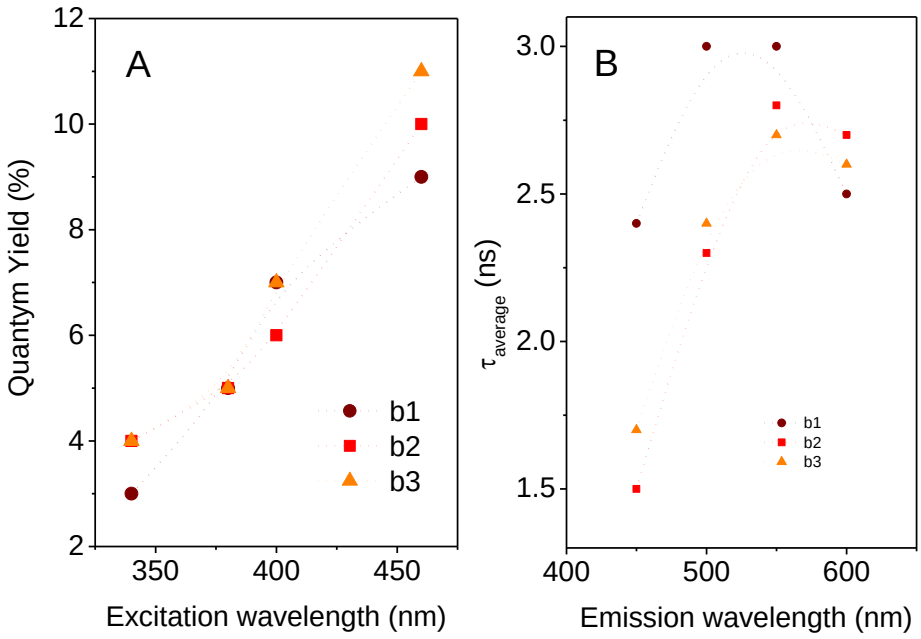

Figure S3. Excitation wavelength dependence of the quantum yield (A) and emission wavelength dependence of the average lifetime (B).

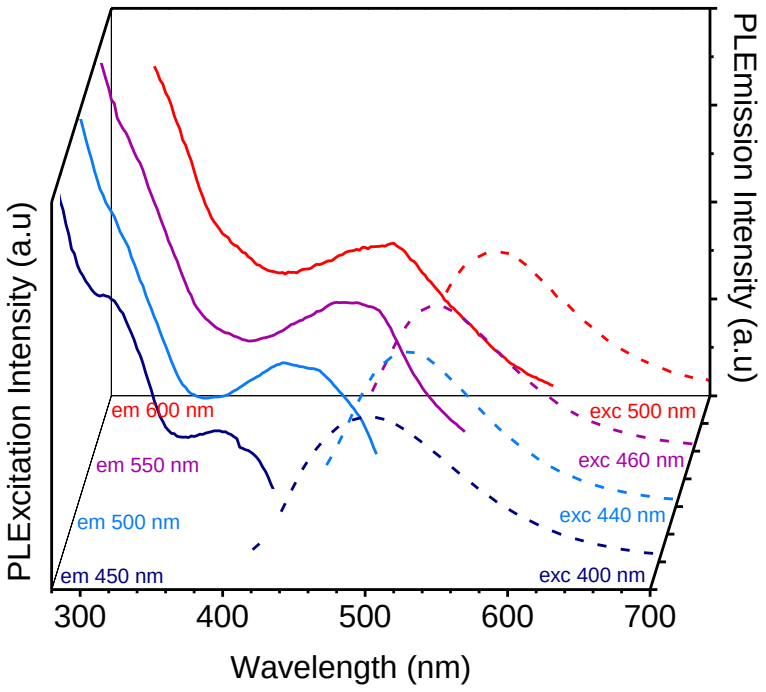

Figure S4. Emission spectrum upon excitation at different wavelengths and photoexcitation spectrum collected at different emission wavelength for b3.

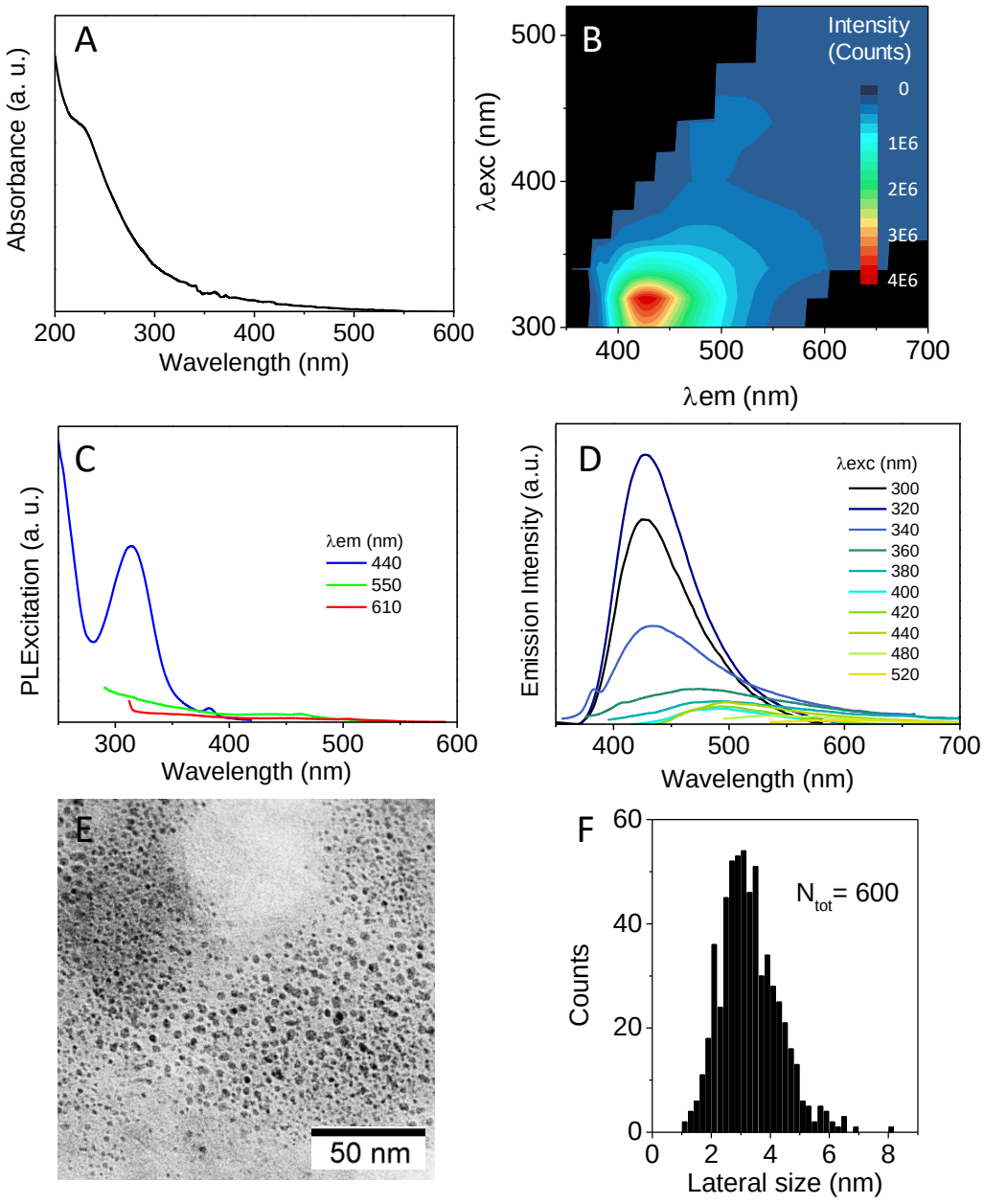

Figure S5. Optical properties and size distribution of undoped dots. (A) Absorption spectrum; (B) contour plot showing of the λexc *vs* λem; (C) photoexcitation spectra collected at different emission wavelength (λem), (D) representative TEM image of the undoped dots and (F) size distribution based on the Ferret diameter measured in the TEM images.

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